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# A REVIEW OF NANOCELLULOSE SYNTHESIS METHODS AND ITS APPLICATION

## Review Metode Sintesis Nanoselulosa dan Aplikasinya

**Fuad Dimar Fauzi, Hafidz Dhany Wicaksono, Joko Waluyo***
Department of Chemical Engineering, Sebelas Maret University, Halilintar Street no. 52, Jebres Jebres District, Surakarta 57126, Central Java, Indonesia
*Email: jokowaluyo@staff.uns.ac.id

***ABSTRACT***
*Nanocellulose is a type of cellulose that is being widely developed to replace petroleum-based polymers. This material possesses biocompatible properties, is abundant in nature, and is eco-friendly due to its biodegradability, sustainability, and non-toxic nature. Various nanocellulose synthesis methods are employed, including acid hydrolysis, alkaline, mechanical and biological treatments, as well as ionic liquid and deep eutectic solvent methods. The choice of synthesis method greatly influences the particle size and crystallinity of the resulting nanocellulose; hence further investigation is needed to determine the effectiveness of these methods. Nanocellulose finds applications in various fields, such as films, polymers, cosmetics, medical fuels, and energy storage. Among the different nanocellulose synthesis methods, ionic liquid and deep eutectic solvent (DES) methods have environmentally safe waste with better temperature, time, and diameter control compared to other methods. However, the DES method is currently preferred over the ionic liquid method due to the possibility of separating the lignin waste from the solvent*.

***Keywords****: Deep eutectic solvent*, *film, ionic liquid*, *nanocellulose, polymer*

**ABSTRAK**
Nanoselulosa merupakan material selulosa yang sedang dikembangkan oleh para peneliti untuk menggantikan polimer berbasis minyak bumi. Material ini memiliki sifat biokompatibel, terdapat dalam jumlah melimpah di alam, serta ramah lingkungan disebabkan oleh sifatnya yang mudah terdegradasi, tidak beracun, dan dapat diperbarui. Metode sintesis nanoselulosa yang umum digunakan meliputi hidrolisis asam, pelarut alkali, mekanik, dan biologi serta metode *ionic liquid* dan *deep eutectic solvent.* Teknik-teknik sintesis ini sangat berpengaruh terhadap ukuran partikel dan kristalinitas nanoselulosa yang dihasilkan, sehingga diperlukan pendekatan lebih lanjut untuk mengetahui efektifitas dari berbagai metode yang digunakan. Secara umum, hasil studi terhadap nanoselulosa menunjukkan bahwa material ini memiliki banyak kegunaan, seperti pada proses pembuatan film, polimer, kosmetik, bidang medis, bahan bakar, dan material penyimpanan energi. Dari berbagai metode sintesis nanoselulosa, metode *ionic liquid* dan DES memiliki limbah aman yang ramah lingkungan dengan suhu, waktu, dan diameter yang lebih baik dari metode lainnya. Namun, metode DES lebih banyak digunakan pada saat ini daripada metode *ionic liquid* karena lignin hasil ekstraksi pada metode DES dapat dipisahkan dari pelarutnya.

**Kata kunci**: *Deep eutectic solvent*, *film*, *ionic liquid*, nanoselulosa, polimer

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## INTRODUCTION

In the past two decades, polymer synthesis has caused significant environmental problems, including plastic waste accumulation, global climate change, and the depletion of fossil fuel reserves. These challenges have driven researchers and industries to focus on the production of bio-polymers from renewable resources as a substitute for petroleum-based polymers (Kumar and Thakur 2017).

Cellulose, one of the most abundant natural polymers, has emerged as a widely used bio-polymer. It consists of glucose bonds arranged in a linear chain, with the C-1 of each glucose molecule binding to the C-4 of the adjacent glucose molecule (Effendi et al. 2015). The linear structure of cellulose contributes to its crystalline nature, making it relatively insoluble in water. In nature, cellulose is found in the form of lignocellulose, which comprises cellulose and other compounds such as hemicellulose, lignin, pectin, and wax. Lignocellulosic materials can be obtained from various agricultural sources, including sugarcane, rice straw, palm fruit, and sago pulp. Nanocellulose, a type of cellulose, is being extensively studied and utilized due to its biocompatible properties, abundant availability, and eco-friendliness. These advantages stem from its biodegradability, sustainability, and non-toxic nature (Mulyadi 2019).

The utilization of cellulose in the form of nanocellulose has gained significant attention. Several techniques can be employed to obtain nanocellulose, including mechanical, chemical, and enzymatic methods. To ensure successful synthesis, characterization of the resulting nanocellulose is essential. This characterization can include size/scale dimension, specific strength, modulus, and surface area analysis (Triyastiti and Krisdiyanto 2017).

Nanocellulose is classified into several categories (Table 1). These comprise nanocrystalline cellulose (CNC), cellulose nanofibrils (CNF), and bacterial nanocellulose (BNC). In each category, nanocellulose has unique characteristics and different morphological structures. For instance, CNC possesses a rod-like structure with diameters in the range of 3-20 nm and lengths in the range of 100-3,000 nm, while BNC has a ribbon-like structure with diameters in the range of 30-100 nm and lengths of 200-3,000 nm (Hitam and Jalil 2022).

Nanocellulose can be prepared from organic materials that are widely found in marshland, including swamp plantations/weeds or agricultural waste, such as rice husk, rice straw, corn straw, midin, melastomes, weeping paperbark, bamboo, palm kernel meal, palm leaf, palm fronds, coconut shell, and water chestnut. These materials were characterized for their cellulose, hemicellulose, and lignin contents, as summarized in Table 2.

## METHODS

There are various methods to synthesize nanocellulose, including acid hydrolysis, alkaline solvation, biological, mechanical, ionic liquid, and deep eutectic solvent (DES) methods. Acid hydrolysis, alkaline solvation, ionic liquid, and deep eutectic solvent are chemical methods. Meanwhile, the mechanical methods comprise ultrasonication- and microwave-assisted treatments.

### Acid hydrolysis method

Acid hydrolysis generally employs strong acids. Strong acids can eliminate the amorphous regions of the cellulose chains (Oke 2010). The mechanism of strong acid hydrolysis is depicted in Figure 1.

In the synthesis of nanocellulose, sulfuric acid ($H_2SO_4$) is a commonly used strong acid. A solution of $H_2SO_4$ 65% was employed by Arjuna et al. (2018) to transform cabbage waste into

**Table 1.** Classification of nanocellulose

| Parameters | Cellulose Nanocrystal (CNC) | Cellulose Nanofibril (CNF) | Bacterial Nanocellulose (BNC) |
|---|---|---|---|
| Physical form | Rod-like | Fiber | Ribbon-like |
| Diameter (nm) | 3-20 | 5-60 | 30-100 |
| Length (nm) | 100-350 | 50-3000 | 200-3000 |

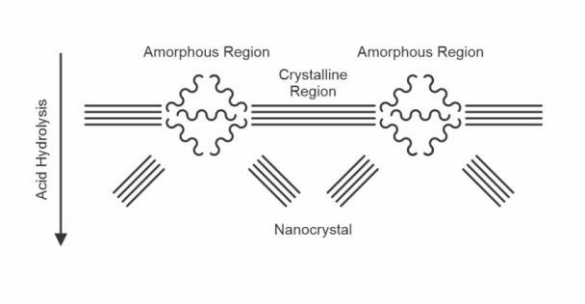


**Figure 1.** Acid hydrolysis process

nanocellulose. The hydrolysis process was carried out at a temperature of 45 °C for 45 min. Prior to hydrolysis, the cabbage waste was delignified in 400 mL of 4% sodium hydroxide solution (NaOH) for 4 h, followed by bleaching in 300 mL of 1.7% sodium hypochlorite (NaOCl) mixed with 100 mL of acetic acid buffer with pH 3 and 100 mL of distilled water for 30 min. Subsequently, the separation process between $H_2SO_4$ and cabbage waste fibers after hydrolysis was performed in a centrifuge at a rotation speed of 3,000 rpm for 5 min until a neutral pH was attained. The characterization results showed that the increase in the crystallinity index of the nanocellulose was 62.27% after bleaching and 15.74% after hydrolysis (Arjuna et al. 2018).

Triyastiti and Krisdiyanto (2018) used a 40% $H_2SO_4$ solution to prepare nanocellulose. The process was conducted at a temperature of 50 ºC for 5 hours. Before the hydrolysis, the midrib of the snakefruit tree was isolated using a 3.5% nitric acid ($HNO_3$) solution and sodium nitrite ($NaNO_2$) at a temperature of 90 ºC for 2 hours (Triyastiti and Krisdiyanto 2018). In another study, Fitriana et al. (2018) also utilized a 60% $H_2SO_4$ solution at a temperature of 60 ºC for 1 hour. Prior to the hydrolysis process, an alkaline treatment was performed using a 1 M NaOH solution at a temperature of 80 ºC for 2 hours. Afterward, the bleaching process was conducted using a 24% hydrogen peroxide ($H_2O_2$) solution at a temperature of 70 ºC for 40 minutes (Fitriana et al. 2018). Julianto et al. (2019) employed a 64% $H_2SO_4$ solution at a temperature of 40 ºC for 45 minutes. Before the hydrolysis, a bleaching process was carried out to remove lignin and hemicellulose fibers in oil palm empty fruit bunches, resulting in a reduction in the fiber diameter. After the hydrolysis, the cellulose was neutralized to achieve pH 7 using a NaOH solution. The TEM images indicated a single fiber at 80,000x magnification with a diameter of 5.1-9.1 nm (Julianto et al. 2017).

In addition to $H_2SO_4$, hydrochloric acid (HCl) is also used to synthesize nanocellulose. This was verified by a study conducted by Maryam et al. (2019), who used a 5 M HCl solution to convert coconut water waste into nanocellulose. The process was carried out at a temperature of 55 ºC for 24 hours. Before the hydrolysis, the coconut water waste from coconut milk processing was reduced and dried to obtain bacterial cellulose (nata de coco) with a diameter of 100 mesh (Maryam et al. 2019).

The acid hydrolysis method was also employed to enhance the bioconversion of biomethane using a diluted acetic acid ($CH_3COOH$) solution. After pretreatment with a 2% (v/v) $CH_3COOH$ solution for 24 hours, the cellulose and hemicellulose content were

**Table 2.** Characterization results of organic materials for nanocellulose synthesis

| No | Material | Content (%) | | | Ref. |
|---|---|---|---|---|---|
| | | Cellulose | Hemicellulose | Lignin | |
| 1. | Melastomes (*Melastomataceae*) | 50.88 | 20.58 | 27.71 | |
| 2. | Midin (*Stenochlaena palustris*) | 42.67 | 21.15 | 35.76 | |
| 3. | Corn straw (*Zea mays*) | 34.06 | 37.63 | 17.03 | |
| 4. | Palm leaf (*Elaeis sp.*) | 42.93 | 26.33 | 30.08 | |
| 5. | Rice husk (*Oryza sativa*) | 43.46 | 23.16 | 32.69 | |
| 6. | Chinese waterchesnut (*Eleocharis dulcis*) | 43.82 | 19.17 | 26.34 | |
| 7. | Rice straw (*Oryza sativa*) | 40.43 | 30.72 | 24.49 | (Maftu et al. 2015) |
| 8. | Palm bunch (*Elaeis sp.*) | 49.63 | 20.69 | 25.60 | |
| 9. | Bamboo (*Bambusa vulgaris*) | 50.11 | 24.57 | 24.81 | |
| 10. | Palm fronds (*Elaeis sp.*) | 50.14 | 24.39 | 25.45 | |
| 11. | Coconut shell (*Cocos nucifera*) | 46.36 | 19.65 | 31.87 | |
| 12. | Palm kernel meal (*Elaeis sp.*) | 51.56 | 14.04 | 34.45 | |
| 13. | Weeping paperbark (*Melaleuca leucandra*) | 51.03 | 23.32 | 34.45 | |
| 14. | Corncob (*Zea mays*) | 40-60 | 20-30 | 15-30 | (Ariyantini 2017) |

**Table 3.** Raw materials, methods, and synthesis products using acid hydrolysis method

| No | Raw Material | Method | Nanocellulose Products | Ref. |
|---|---|---|---|---|
| 1. | Cabbage waste | 1. Delignification, NaOH<br>2. Bleaching, NaClO<br>3. Acid hydrolysis, $H_2SO_4$ | Nanocellulose<br>- CI : 78,01%<br>- D : 58,91 nm | (Arjuna et al. 2018) |
| 2. | Midrib of snakefruit tree | 1. Nanocellulose isolation, $HNO_3$ + $NaNO_2$<br>2. Acid hydrolyis, $H_2SO_4$ | Nanocellulose<br>- CI : 58.42%<br>- D : 16.52 nm | (Triyastiti and Krisdiyanto 2018) |
| 3. | Bamboo shoots | 1. Alkaline treatment, NaOH<br>2. Bleaching, $H_2O_2$<br>3. Acid hydrolysis, $H_2SO_4$ | - | (Fitriana et al. 2018) |
| 4. | Oil palm empty fruit bunches | 1. Bleaching<br>2. Acid hydrolysis, $H_2SO_4$ | Nanoparticles<br>- D : 5.1-9.1 nm | (Julianto et al. 2017) |
| 5. | Coconut waste water from coconut milk production (santan) | 1. Synthesis of bacterial cellulose (nata de coco)<br>2. Acid hydrolysis, HCl | Cellulose<br>- D : 535.8 nm | (Maryam et al. 2019) |
| 6. | Rice straw | 1. Acid pretreatment | - | (Peng et al. 2019) |

Note: CI = Crystal Index, D = Diameter

significantly reduced by 8.9% and 18.1% respectively, at 80 ºC with a liquid to solid ratio of 1:10 (v/w). In this study, the concentration of acetic acid only had a slight influence on the conversion compared to the reaction time, which significantly affected the extraction of lignocellulose from the straw. Additionally, the loss of lignin was around 3-4.7% (Peng et al. 2019).

The acid hydrolysis method enables the production of nanocellulose with a fairly high crystallinity index. However, strong acids have negative impacts due to their toxic, dangerous, and corrosive nature, requiring careful handling. Moreover, strong acid waste must be treated before being disposed of in the environment to avoid pollution (Effendi et al. 2015).

## Alkaline solvation method

The alkaline treatment method, also known as mercerization, is performed by treating cellulose fibers with strong bases such as NaOH or KOH (Dhali et al. 2021). Alkaline solvation treatment can be divided into two methods: heating the alkaline solution and alkaline cooking using a digester or autoclave. According to Ng et al. (2015), the heating method involves maintaining the alkaline solvent at a

**Table 4.** Raw materials, methods, and synthesis products using alkali solvents method

| No | Raw Material | Method | Nanocellulose Products | Ref. |
|---|---|---|---|---|
| 1. | Rice straw | 1. Alkaline pretreatment, NaOH | - | (Hang Shu et al. 2015) |
| 2. | Cassava stem | 1. Pretreatment<br>2. Delignification, $Na_2SO_3$<br>3. Bleaching, $H_2O_2$, | - | (Lismeri et al. 2016) |
| 3. | Pineapple fiber | 1. Delignification, NaOH<br>2. Bleaching, NaOCl<br>3. Acid hydrolysis, $H_2SO_4$ | Nanocellulose<br>- CI : 76.5%<br>- D : 134-407 nm | (Evelyna et al. 2019) |
| 4. | Onion peels | 1. Delignification, $C_2H_5OH$<br>2. Extraction, $H_2O_2$ + NaOH<br>3. Preparation, $H_2SO_4$ | Nanofibril<br>- CI: 78.668%<br>- D : 12.615 nm | (Hertiwi et al. 2020) |
| 5. | Ambon banana peel waste | 1. Cellulose extraction, NaOH<br>2. Bleaching, NaOCl<br>3. Cellulose isolation, $(NH_4)_2S_2O_8$ | Nanocellulose<br>- CI : 60.89-72.40%<br>- D : 12.1-25.4 nm | (Nugraha et al. 2021) |

temperature of 70 °C to 90 °C, while alkaline cooking is carried out at a temperature of 160 °C under high pressure.

The main objective of alkaline solvation is to improve the probability of depolymerization and solvation of lignin and branched hemicellulose hydrolysis, which releases cellulose content and facilitates subsequent processing stages (Maurya et al. 2015). However, it is crucial to control the alkaline solvent treatment to avoid undesirable cellulose degradation or hindered reactions (Maurya et al. 2015). Based on literature studies, Table 4 provides a summary of research investigating the extraction of nanocellulose using the alkaline solvation method.

The alkaline solvation method is often employed after acid hydrolysis treatment to eliminate the hemicellulose content in lignocellulosic materials (Sanchez et al. 2011). NaOH and sodium sulfite ($Na_2SO_3$) can be used as alkaline solvents for the synthesis of nanocellulose, with NaOH being the most commonly used base. Evelyna et al. (2019) employed a 4 M NaOH solution to prepare nanocellulose from pineapple fiber at a temperature of 80 °C for 4 hours. The pineapple fiber was bleached using 5% NaOCl, followed by acid hydrolysis using 45% $H_2SO_4$ at a temperature of 45 °C for 90 hours (Evelyna et al. 2019).

In another study, Hertiwi et al. (2020) also implemented the alkaline solvation method using a 4% NaOH solution at a temperature of 60 °C for 2 hours. Prior to the extraction process, the onion peel was delignified with an ethanolic solution with a 1:1 (v/v) ratio of ethanol to water. Subsequently, the sample preparation using 50% $H_2SO_4$ was conducted until suspension in the solution was formed, followed by characterization (Hertiwi et al. 2020). Nugraha et al. (2021) utilized a 1 M NaOH solution at a temperature of 80 °C for 4 hours. The bleaching process was then performed using a 1.75% NaOCl solution at a temperature of 70 °C for 1 hour, followed by nanocellulose isolation using a 1 M ammonium peroxodisulfate ($(NH_4)_2S_2O_8$) solution within a temperature range of 60-80 °C for 16 hours (Nugraha et al. 2021).

Aqueous sodium sulfite ($Na_2SO_3$) solution with a concentration of 20% (w/w) was used in a study carried out by Lismeri et al. (2016) at 100 °C for 2-3 hours. Prior to the delignification process using an alkaline solvent, the cassava stem was pretreated using an acidic solution with a 1:6 (w/w) ratio at a temperature of 90 °C for 1 hour. Subsequently, the delignification process was performed by bleaching with an $H_2O_2$ solution at room temperature for 3 hours. The results indicated that the pretreatment triggered the disruption of lignin and hemicellulose structures. Additionally, the preparation and the ratio of materials to the solvent used throughout the experiments influenced the degradation of lignin (Lismeri et al. 2016).

The alkaline solvent was also utilized in the production of biomethane, according to a study conducted by Hang Shu et al. (2015). The pretreatment of rice straw was performed to produce methane using a 1.5 M NaOH solution with a solid-to-liquid ratio of 1:10, kept at 30 °C for 5 hours. From the experiments, treatment with the alkaline solvent altered the lignin content from 10.64% to 5.8%, cellulose content from 38.2% to 81.1%, and hemicellulose content from 30.8% to 8.23% (Hang Shu et al. 2015).

The drawbacks of the alkaline treatment method emerge during the solvation process. In this process, the glycosidic bond is broken down to break the lignin structure. Consequently, the lignin cannot preserve the cellulose during the dissolution process, leading to the formation of cellulose monomers (glucose).

**Biological Methods**

Nanocellulose can be synthesized using biological methods with the help of bacteria. The one that is frequently used is *Acetobacter xylinum*. *A. xylinum* is a type of gram-negative bacteria that is able to produce cellulose and acetic acid during its growth. The resulting cellulose subsequently forms a dense form of fibrils known as nata. At the time of his death, Felasih et al. (2010) applied biological methods using *A. xylinum*. *A. xylinum* is grown in coconut water as its medium. The cellulose production is carried out at a temperature of 30 ± 2 °C for 14 days (Felasih et al. 2010).

Nanocellulose production using *A. xylinum* in the medium of coconut water has also been done by Ifadah et al. (2015) within 14 days (Ifadah et al. 2015). Afterwards,

Wibowo et al. (2016) used coconut water, which contains 91.2% water, 0.29% protein, 0.15% fat, 7.27% carbohydrates, and 1.06% ash, to grow *A. xylinum*. *A. xylinum* produces extracellular enzymes that are capable of polymerizing sugar (glucose) into thousands of cellulose chains (Wibowo and Isroi 2016).

In addition to using coconut water, *A. xylinum* can also grow on pineapple peel extract medium at a temperature of 30 °C for 14 days (Muhajir et al. 2018) and in sago liquid waste medium with the addition of granulated sugar as the source of nitrogen and glacial acetic acid to regulate the pH to about 4-5 (Ahmad et al. 2019).

Besides *A. xylinum*, *A. niger* can also be used to synthesize bacterial nanocellulose. The same research by Hang Shu et al. (2015) used *A. niger* with the same material but applied biological pretreatment. This involved adding straw to an autoclave at a temperature of 121 °C for 20 minutes, followed by adding *A. niger* to it at a temperature of 30 °C for 5-15 days. The results obtained after 5 days of the experiment showed a reduction of lignin from 10.64% to 10.43%, cellulose from 38.2% to 37.29%, and hemicellulose from 30.8% to 30.6% (Hang Shu et al. 2015).

The biological method has a drawback, which is the difficulty in obtaining certain enzymes in a pure state. Additionally, the time required to obtain cellulose tends to be longer compared to chemical methods.

**Mechanical method**

According to Phanthong et al. (2018), the mechanical method is a process of isolating cellulose fibers by applying high shear force to separate cellulose fibers longitudinally, which will produce cellulose nanofibrils (CNF). The commonly used approaches in the mechanical method are high-pressure homogenization (HPH), ultrasonication, and ball grinding (Phanthong et al. 2018).

High-pressure homogenization (HPH) occurs when the cellulose slurry passes into the high-speed and high-pressure process vessel (Abdul Khalil et al. 2014). Ultrasonication is a process of defibrillating cellulose fibers with hydrodynamic force caused by ultrasound. In this process, mechanical isolation forces are applied, resulting in the formation, expansion, and bursting of microscopic gas bubbles when liquid molecules absorb ultrasonic energy (Zhou et al. 2012). Another mechanical method that can defibrillate cellulose is ball grinding. This method causes a shear force between the balls and the surface of the container, resulting in cellulose fibers with a smaller diameter due to cracks in the cellulose structure (Kim et al. 2013).

In their study, Saputri and Sukmawan (2020) used the ultrasonication method to obtain nanocellulose. The preparation involved cutting, drying, smoothing, and then shrinking the sample with a grinder, followed by sieving to obtain a water content of 4-5%. Subsequently, there was a washing and stirring step at a temperature of 80 °C for 1 hour, followed by another wash with cold water. Delignification was carried out using a 5% NaOH solvent at a temperature of 80 °C and a speed of 250 rpm for 3 hours. Before

**Table 5.** Raw materials, methods, and synthesis products using biological method

| No | Staple | Method | Nanocellulose Result | Reference |
|---|---|---|---|---|
| 1. | Coconut Water | Enzymatic, *A. xylium* | Bacterial nanocellulose<br>- h: 0.09-0.1 mm | (Felasih 2010) |
| 2. | Coconut Water | Enzymatic, *A. xylium* | Bacterial nanocellulose<br>- h : 4.37-8.76 mm | (Ifadah et al. 2015) |
| 3. | Rice straw | Enzymatic, *Aspergillus niger* | - | (Hang Shu et al. 2015) |
| 4. | Coconut Water | Enzymatic, *A. xylium* | Bacterial nanocellulose | (Wibowo and Isroi 2016) |
| 5. | Pineapple Peel | Enzymatic, *A. xylium* | Bacterial nanocellulose<br>- CI: 74-83%<br>- D : 0.99-1.05 nm | (Muhajir et al. 2018) |
| 6. | Sago Liquid Waste | Enzymatic, *A. xylium* | Bacterial nanocellulose<br>- h : 21.8 mm | (Ahmad et al. 2019) |

**Table 6.** Raw materials, methods, and synthesis products using mechanical method

| No | Staple | Method | Nanocellulose Result | Reference |
|---|---|---|---|---|
| 1. | Palm trunk | 1. Preparation<br>2. *Pre-treatment*<br>3. Delignification, NaOH<br>4. Oxidation, NaOH<br>5. *Bleaching*, $H_2O_2$<br>6. *Blending*<br>7. Ultrasonication | - | (Saputri and Sukmawan 2020) |
| 2. | Microcrystalline cellulose (MCC) | 1. Ultrasonication, $H_2SO_4$<br>2. *Microwave*<br>3. Centrifugation<br>4. Purification | - | (Kos et al. 2014) |
| 3. | Bagasse | 1. *Bleaching*, NaOH<br>2. Extraction, distilled water + $H_2O_2$ + NaOH<br>3. *Blending* | - | (Saputri and Sukmawan 2020) |
| 4. | Wild bushes | 1. Acid hydrolysis, $H_2SO_4$<br>2. Ultrasonication | Nanocellulose<br>- D : 1,223-3,502 µm | (Widiastuti and Marlina 2020) |
| 5. | Snake fruit midrib | 1. Acid hydrolysis<br>2. Ultrasonication | Nanocellulose<br>- CI: 70.4%<br>- D: <100 nm | (Yudha 2018) |

bleaching, the NaOH solvent was oxidized to reach pH 11, and then the sample was bleached using $H_2O_2$ at a temperature of 80 °C for 2 hours. The next step involved blending it at 21,000 rpm for 10 minutes, followed by ultrasonication using an ultrasonic cell crusher for 10 minutes. The test results showed that the raw fiber still contained a mixture of crystalline and amorphous structures. After the bleaching process, the amorphous content was slightly reduced. Then, after the blending process, the fiber became more organized and shorter, while after ultrasonication, the fiber became smaller, and the crystalline structures became more visible (Saputri and Sukmawan 2020).

Mechanical method uses a microwave to synthesize nanocellulose. First, microcrystalline cellulose, up to 0.5 g, is added with 10 g $H_2SO_4$ with varying concentrations as listed. The formed suspension is then ultrasonicated for 5 minutes. The sample is heated to the listed temperature for 1 minute and kept constant for 10 minutes. After that, it is cooled at a temperature of 50 °C for 2 minutes. During the process, each variation is stirred at a speed of 600 rpm. Once the reaction is complete, remove the sample from the microwave and cool it under a tap water until it reaches room temperature. The cooled suspension is later transferred to a 15 mL plastic tube and centrifuged at a speed of 4700 rpm for 10 minutes to remove excess acid and water. The precipitate formed is purified by washing it 5 times with 5 × 50 mL deionized water and repeating the ultrasonication process for 5 minutes, followed by grinding (centrifugation) at a speed of 9000 rpm for 20 minutes. The final purification step involves sonication for 30 minutes with a power of 230 W $cm^{-2}$, and the obtained pH after ultrasonication is between 4 to 5. The formed nanocrystals are determined gravimetrically by drying the precipitate after shaking it at a speed of 8000 rpm under vacuum conditions at a temperature of 40 °C for 10 minutes. The best results were obtained at the concentration of $H_2SO_4$ with a crystal index of 68-82% (Kos et al. 2014).

The mechanical method uses a blender to synthesize nanocellulose. As presented by Saputri and Sukmawan (2020), before blending the bagasse, it is first bleached using a 5% NaOH solvent. Furthermore, extraction is carried out to remove lignin using a mixture of distilled water, $H_2O_2$, and NaOH, and then the blending process is conducted. From the blending results, the sample with a concentration of 0.7% contains more nanocellulose than others, with a high degree of crystallinity (Saputri and Sukmawan 2020).

In a study by Widiastuti and Marlina (2020), the mechanical method of ultrasonication is performed for 20 minutes. Before sonication is conducted, the $H_2SO_4$ 64% solvent is bleached first. The result of this process is in the form of nanocellulose with a diameter of 1.223-3.502 (Widiastuti and Marlina 2020). Other ultrasonication mechanical methods were also carried out in studies conducted by Yudha et al. (2018). Before sonication is done, the snake fruit midrib is hydrolyzed with acid first. The results of this process are nanocellulose with a crystal index of 70.4% and a diameter of <100 nm (Yudha, 2018).

Mechanical methods have the disadvantage of high production costs, including tools and materials. Additionally, this method requires more energy compared to chemical methods.

**Ionic liquid method**

The ionic liquid method is utilized for the synthesis of nanocellulose by dissolving cellulose through the disruption of intermolecular hydrogen bonds. Ionic liquids are classified as non-derivatization solvents and their ability to dissolve cellulose depends on the size and polarity of their anions and cations. The cation in the ionic liquid replaces the oxygen atom of the cellulose-OH group, while the anion replaces the hydrogen atom of the cellulose-OH group (Wang et al. 2017).

Research has shown that chloride anion ($Cl^-$) can dissolve cellulose more effectively than other anions (Swatloski et al. 2002). In the study conducted by Wang et al. (2017), the $Cl^-$ anion was paired with the 1-butyl-3-methyl-imidazolium cation ($[Bmim]^+$) using the ionic liquid [Bmim]Cl to dissolve cellulose from bleached bagasse. This process was carried out at a temperature of 130 °C with the assistance of a microwave for 2 hours. Similarly, Phanthong et al. (2017) used the ionic liquid [Bmim]Cl to dissolve cellulose from cellulose powder. This process was performed at room temperature using ball milling at a speed of 400 rpm for 2 hours. Additionally, $Cl^-$ anions can also be combined with [Emim] and [Pmim] to form ionic liquids for the dissolution of cellulose from microcrystalline cellulose (MCC). This process was conducted at a temperature of 100 °C for 12 hours (Babicka et al. 2020).

Apart from the $Cl^-$ anion, [Bmim] can also be combined with other anions to form 1-butyl-3-methyl-imidazolium acetate ([Bmim]OAc) and 1-butyl-3-methyl-imidazolium tetrafluoroborate ([Bmim]BF4). Abushammala et al. (2015) used the ionic liquid [Bmim]OAc to dissolve cellulose from wood flower at a temperature of 60 °C for 4 hours, while Eksiler et al. (2017) employed the ionic liquid [Bmim]BF4 to dissolve cellulose from Palm mesocarp at room temperature for 3 hours (Abushammala et al. 2015; Eksiler et al. 2017).

Then, Asim et al. (2021) conducted delignification testing using the acid ionic liquid (AIL) method. In the aforementioned

**Table 7.** Raw materials, methods, and synthesis products using ionic liquid method

| No | Staple | Method | Nanocellulose Result | Reference |
|---|---|---|---|---|
| 1. | *Wood flower* | *Ionic liquid*, [Bmim] OAc, | Nanocrystal<br>- D: 2-5 nm<br>- CI: 75% | (Abushammala et al. 2015) |
| 2. | *Bleached bagasse* | *Ionic liquid*, [Bmim] Cl | Nanoparticles<br>- D : 10-20 nm<br>- CI : 36% | (Wang et al. 2017) |
| 3. | Cellulose powder | *Ionic liquid*, [Bmim] Cl | Nanocrystal<br>- D : 10-25 nm<br>- CI : 66% | (Phanthong et al. 2017) |
| 4. | Palm trunk mesocarp | *Ionic liquid*, $[Bmim]BF_4$ | Nanoparticles<br>- D : 127 nm<br>- CI : 27% | (Eksiler et al. 2017) |
| 5. | *Microcrystalline Celullose (MCC)* | *Ionic liquid*, [Emim] CL and [Pmim] Cl | *Sigmacell*<br>- D : 100 nm | (Babicka et al. 2020) |
| 6. | Rice straw | *Ionic liquid*, [PyH] $[HSO_4]$, [PyH] $[HSO_4.H2SO_4)]$, and [PyH] $[HSO_4 .H_2SO_4)_3]$ | - | (Asim et al. 2021) |

study, three protic ionic liquids based on pyridinium cation and hydrogen sulfate anion ($HSO_4^-$) were synthesized: [PyH] [HSO4] (IL1), [PyH] [$HSO_4.H_2SO_4$)] (IL2), and [PyH] [$HSO_4.H_2SO_4)_3$] (IL3), and were used to isolate lignocellulose in straw. From the conducted experiments, the most effective ionic liquid in terms of lignin removal was the compound IL3, characterized by a high delignification rate (79%) and lignin recovery (77%) at a temperature of 60 °C for 2 hours (Asim et al. 2021).

In contrast to molten salt, ionic liquids generally have high viscosity and melting points, making them corrosive. However, at room temperature, ionic liquids are typically in a liquid state and have relatively low viscosity, making them non-corrosive (Stolte et al. 2006). Ionic liquids can dissolve whole cellulose biomass or selectively remove lignin and hemicellulose (Elgharbawy et al. 2016). Although ionic liquids are considered "green solvents," further studies on their toxicity towards enzymes and microorganisms should be conducted, and low-cost technologies for recovery and reuse should be carefully considered before their application (Dhali et al. 2021).

**Deep eutectic solvent (DES) method**

Deep eutectic solvent (DES) is a new generation of ionic liquids with physical and chemical properties equivalent to ionic liquids (Abbott et al. 2004). This solvent typically consists of two compounds: a hydrogen bond donor and a hydrogen bond acceptor (Le Gars et al. 2020). The interaction between these two compounds leads to eutectic mixtures with lower melting points compared to their individual constituents. Based on the nanocellulose surface, DES can be categorized into two types: DES derivative and DES non-derivative (Jiang et al. 2021). DES offers several advantages over the ionic liquid method, including affordability, non-flammability, non-toxicity, thermodynamic stability, and biodegradability (Liu et al. 2017; Wang et al. 2020b). Furthermore, studies have shown that pre-treatment with DES from lignocellulosic biomass facilitates easier lignin recovery while preserving the integrity of cellulose, making it suitable for producing high-value chemical compounds (Francisco et al. 2012; Alvarez-Vasco et al. 2016; Tang et al. 2017; Kuang et al. 2018; Das et al. 2018; Wang et al. 2020a).

A study by Sirviö et al. (2015) utilized DES method to synthesize nanocellulose from bleached birch pulp. Choline chloride was used as the acceptor, while urea served as the donor. The synthesis was conducted in a round-bottom flask at a temperature of 100 °C for 15 minutes, resulting in nanocellulose with a diameter of 2-5 nm (Sirviö et al. 2015). In another experiment conducted by Li et al. (2018), DES method was employed to synthesize nanocellulose from dialdehyde cellulose. Aminoguanidine hydrochloride was used as the acceptor, and

**Table 8.** Raw materials, methods, and synthesis products using deep eutectic solvent (DES) method

| No | Staple | Method | Nanocellulose Result | Reference |
|---|---|---|---|---|
| 1. | Bleached birch pulp | DES, cholin chloride-urea | Nanofibril<br>- D: 2-5 nm | (Sirviö et al. 2015) |
| 2. | Dialdehyde cellulose | DES, aminoguanidine hydrochloride-glycerol | Nanofibril<br>- D : 4.6-5.7 nm<br>- CI : 64% | (Li et al. 2018) |
| 3. | Wheat | DES, cholin chloride-levulinic acid | Nanofibril<br>- D: 4-6 nm<br>- CI: 38-54% | (Suopajärvi et al. 2020) |
| 4. | Softwood pulp | DES, ammonium formate-organic acid | Nanocrystal<br>- L : 100-300 nm<br>- D : 10 mm | (Jaekel et al. 2021) |
| 5. | Dissolving pulp/microcrystalline cellulose | DES, guanidine hydrochloride-phosphoric acid | Nanoparticles<br>- D : 5.6-5.8 nm | (Sirviö 2019) |
| 6. | Sorghum straw | DES, ChCl-EG, ChCl-UR, ChCl-GLY, ChCl-LAC, ChCl-MA, ChCl-CA, 150 °C | - | (Wu et al. 2021) |

glycerol was used as the donor. The synthesis was carried out at a temperature of 70 °C for 10 minutes, yielding nanocellulose with a crystal index of 64% and a diameter of 4.6-5.7 nm (Li et al. 2018).

The development of nanocellulose extraction by the DES method does not stop there. Subsequent research was conducted by Suopajärvi et al. (2019) using DES method to synthesize wheat into nanocellulose. Choline chloride is used as an acceptor, while levulinic acid is used as a donor. The synthesis is carried out at a temperature of 70 °C for 10 minutes. The results obtained are in the form of nanocellulose with a crystal index of 38-54% and a diameter of 4-6 nm (Suopajärvi et al. 2020). Another study was conducted by Jaekel et al. (2021) using DES method to synthesize nanocellulose. Ammonium formate is used as an acceptor, while organic acids such as glycolic, lactic, and levulinic acid are used as donors. The synthesis is carried out inside an autoclave at a temperature of 180 °C for 4 hours. The results obtained are in the form of nanocellulose with a length of 100-300 nm and a diameter of 10 nm (Jaekel et al. 2021).

DES demonstrated the highest effectiveness in terms of producing nanocellulose. DES can be highly effective in selectively removing lignin components while ensuring the integrity of cellulose and hemicellulose fractions (Satlewal et al. 2018). Lignin is the most abundant biopolymer after cellulose, constituting 30% of the organic carbon on Earth (Boerjan et al. 2003). Approximately 50% of lignin components are aromatic hydrocarbons with the potential to be used as fuel and produce aromatic chemical compounds (Xu et al. 2014). Fuels derived from lignin can exist in solid, liquid, or gas forms, such as biochar, transport fuels, or syngas (Tripathi et al. 2016). Numerous studies have employed DES for lignin extraction, and DES acids containing hydrogen bond donor (HBD) acids have demonstrated higher lignin extraction efficiency compared to other DES (Tan et al. 2019). Lignin extraction using DES choline chloride-lactic acid (CC-LA) yields approximately 80.64 ± 1% lignin, while DES choline chloride-formic acid (CC-FA) yields approximately 85.84 ± 1.07% lignin. Lignin CC-FA is suitable as a carbon source due to its thicker structure and higher charcoal residue, whereas lignin CC-LA is recommended as an aromatic raw material for depolymerization processes due to its high β-O-4' bond content and volatility (Tan et al. 2020).

Wu et al. (2021) conducted a study on the improvement of enzymatic saccharification of sorghum straw through a combined delignification method involving pretreatment with alkaline extraction and DES submersion. The material used in this study was sorghum straw. The first pretreatment method involved alkaline extraction using NaOH. Sorghum straw was soaked in a small amount of NaOH solvent (0.75% wt) at an S/L ratio of 1:10 (wt:wt) and autoclaved at 121 °C for 1 hour. Subsequently, DES submersion was carried out by mixing 75 g of DES (ChCl:EG, ChCl:UR, ChCl:GLY, ChCl:LAC, ChCl:MA, or ChCl:CA) with 5 g of sorghum straw at a temperature of 150 °C. In this study, ChCl was used as the hydrogen bond acceptor (HBA), and six different types of hydrogen bond donors (HBD) were employed, namely EG: ethylene glycol, UR: urea, GLY: glycol, LAC: lactic acid, MA: malic acid, and CA: citric acid. Based on the experiments, it can be concluded that the DES pretreatment method is optimal when using ChCl:LAC-SS, which achieved a lignin removal rate of 49%. Furthermore, the DES submersion results with ChCl:LAC-SS were further optimized by combining with NaOH. The parameters used included the S/L ratio (1:10, 1:25, 1:20, 1:25 wt:wt), 5 g of dry sorghum straw pretreated with NaOH extraction mixed with 75 g of DES, with varying temperatures (120, 130, 140, 150, and 160 °C) and duration (20-60 minutes). In this study, the pretreatment using only ChCl:LAC-SS DES immersion was not as effective as the one with alkaline pretreatment. Alkaline delignification (NaOH extraction) resulted in approximately 66.9% cellulose delignification, which significantly differed from the 49% lignin removal achieved by ChCl:LAC-SS DES-based delignification. Therefore, combining alkaline extraction (AE) with DES in the sequence AE-ChCl:LAC-SS and ChCl:LAC-SS-AE produced different percentages of delignification, with each achieving approximately 79.3% and 78.4% delignification, respectively (Wu et al. 2021).

**Table 9.** Comparison of methods

| Parameters | Acid Hydrolysis | Alkali Solvent | Biology | Mechanical | *Ionic Liquid* | DES |
|---|---|---|---|---|---|---|
| Temperature (°C) | 40–60 | 30–100 | 30 ± 2 | – | 30–120 | 30–180 |
| Time (hours) | 3/4–24 | 2–120 | 120–360 | 1/3–1 | 2–12 | 1/6–24 |
| CI (%) | 58.42–78.01 | 60.89–78.67 | 74–83 | 68–82 | 27–75 | 38–64 |
| D (nm) | 5.91–535.8 | 12.1–407 | 0.99–1.05 | <100 | 2–127 | 2–10 |
| Waste | Hazardous | Hazardous | Safe | Hazardous | Safe | Safe |

The hydrolysis process is necessary to separate cellulose and hemicellulose. However, the conventional hydrolysis process involves the use of strong acids and bases that are toxic, and the resulting waste must be treated before disposal. Yang et al. (2020) discovered a method to separate cellulose and hemicellulose using the solvent γ-valerolactone (GVL) (Yang et al. 2020). GVL does not contain harmful compounds, halogens, or phosphorus (Alonso et al. 2013).

Comparing the various methods of producing nanocellulose mentioned above, we can evaluate them based on temperature, time, crystal index (CI), diameter (D), and waste generated.

Based on the comparison table above, the acid hydrolysis, alkali solvent, and mechanical methods produce hazardous waste, which necessitates treatment before disposal. Although biological methods generate harmless waste, the manufacturing process is time-consuming. The ionic liquid and DES methods produce environmentally safe waste, with lower temperature, shorter processing time, and smaller diameter compared to the other methods. The lignin waste obtained from DES extraction can be separated using appropriate solvents and used as an aromatic feedstock, making the DES method more extensively studied than the ionic liquid method to date (Dhali et al. 2021).

## APPLICATION OF NANOCELLULOSE

### Application in film

The application of nanocellulose in film packaging has been explored in several studies. Iriani et al. (2015) investigated the use of nanocellulose as a nanofiller in polyvinyl alcohol-based composite films. Nanocellulose was obtained from pineapple leaf fiber through an acid hydrolysis process using 60% $H_2SO_4$ at a temperature of 45 °C for 60-90 minutes, followed by sonication for 20 minutes. The resulting nanocellulose had an average size of 284.6 nm. Similarly, Chaichi et al. (2017) and Sutay Kocabaş et al. (2021) used cellulose nanocrystals (CNC) as an additional material in edible films for packaging.

Muhajir et al. (2019) investigated the use of nanocellulose as a filler in films. Nanocellulose was obtained from bagasse through an enzymatic process using *Acetobacter xylinum* for 14 days. Aguilar-Sanchez et al. (2021) used CNC and cellulose nanofibers (CNF) as a coating for polyether sulfone (PES) membranes. The results showed a significant decrease in the

**Table 10.** Application of nanocellulose in film

| Application | Material | Result | Reference |
|---|---|---|---|
| Nanofiller film composites for packaging (plastic) | Nanocellulose from pineapple leaf fiber, PVA, and glycerol | The addition of nanocellulose improves the crystallinity properties of PVA-based composite films | (Iriani et al. 2015) |
| Nanocellulose bacteria film filter or sensor application | Pineapple peel waste, *Acetobacter xylinum* | The addition of bacterial nanocellulose increases tensile strength and young modulus of the film | (Muhajir 2019) |
| Bionanocomposite edible film, for food packaging | Pectin and nanocrystal cellulose (CNC) | The addition of 5% CNC concentration increases tensile strength by 84% and decreases water vapor by 40% | (Chaichi et al. 2017) |
| Biodegradable film from bulgur bran, for packaging applications | Bulgur bran and cellulose nanocrystals (CNC), commercial cellulose nanoparticles (CNF) | The addition of 10% CNC and CNF reduces the film water solubility (FWS) by 21.3% compared to the untreated film. | (Sutay Kocabaş et al. 2021) |
| Nanocellulose as a membrane coating polyether sulfone (PES) | Polyethersulfone (PES) and nanocrystal cellulose (CNC) | The membrane coated with CNC or T-CNF shows a significant decrease in contaminating organic compounds, as indicated by a decrease in surface roughness. | (Aguilar-Sanchez et al. 2021) |

contamination of organic compounds, as indicated by a decrease in surface roughness, when the membranes were coated with CNC or T-CNF.

Based on the above examples, the addition of nanocellulose (NC) as a nanocomposite has been shown to improve the physical characteristics of biofilms. The addition of NC improves crystallinity properties, increases tensile strength, reduces water permeation, solubility in water, and prevents contamination by organic compounds. Additionally, the use of nanocellulose in films contributes to environmental sustainability due to its biodegradability.

## Application in polymers

Most of the time, nanocellulose derived from biomass is used in the form of nanocomposites with other materials in polymer applications. The development of bio-composite based materials has gained attention among scientists, with biopolymers such as polylactic acid (PLA), chitosan, cellulose, lignin, etc. being used as bioplastic makers, polymer reinforcing agents, fillers, and recyclable thermoplastics (Marakana et al. 2021).

While food packaging has been a primary application for synthetic polymers, there is now a growing focus on reducing the accumulation of solid waste from synthetic food packaging due to its environmental impact (Geyer et al. 2017). However, one main disadvantage of biopolymers mentioned earlier is their fragility and complexity, making it crucial to incorporate natural fibers as reinforcing components to improve their mechanical properties (Jamr et al. 2019). Cellulose, a natural polymer, holds great promise in this regard. Through various pre-treatment methods, cellulose can be converted into nanocellulose and microcrystalline cellulose (MCC), which can enhance the mechanical resistance of films (Ilyas Rushdana et al. 2017; Mishra et al. 2018). As the demand for efficient and flexible materials increases, there have been several developments and research on the addition of various types of composite films

**Table 11**. Nanocellulose in polymer application

| Application of Nanocellulose | Material | Result | Reference |
|---|---|---|---|
| Addition of nanocellulose for replacing styrofoam | Empty palm bunches | Cellulose produces better biofoam characteristics than nanocellulose. Cellulose modification increases density, tensile strength, and decreases water absorption | (Nurfitasari 2018) |
| Nanocellulose as filler in bioplastic tapioca used for product packaging | Manioc | Addition of nanocellulose increases bioplastic thickness and inhibits light, UV light, and visible light | (Wicaksono et al. 2021) |
| Cotton leaf nanocellulose potential as an injection water viscosity enhancing agent for EOR (Enhanced Oil Recovery) process | Cotton leaf | Cotton leaf nanocellulose increases water viscosity, enhancing the effectiveness and efficiency of the enhance oil recovery method in the petroleum industry | (Khalid et al. 2020) |
| Biocomposite nanocellulose PLA | PLA powder (Matrix) and pulp linter (booster) | The results of mixing the NCF with Maleated PLA increased the characteristic natures of the tensile modulus by 40% and the tensile strength increased from 22.4 MPa to 69.3 MPa from the addition of 5% (w/w) NCF | (Ghasemi et al. 2018) |
| The addition of CNP (Cellulose Nanoparticle) from potato peels on PVA biopolymer to improve the active packaging characteristic | Potato peels | With the addition of CNP, the characteristics of PVA-CNP nanocomposite increase the value of tensile strength and elongation. Therefore, biopolymer active packaging based on the addition of CNP is more | (Ramesh and Radhakrishnan 2019) |

combined with nanocellulose (NC) and MCC.

For example, Nurfitasari (2018) conducted a study comparing the manufacture of biofoam using different types of cellulose from oil palm empty fruit bunches. The best functional characteristics were achieved with cellulose from empty palm bunches (STKS), while nanocellulose caused the biofoam paste to be too thin to improve mechanical properties. The best biofoam was obtained with a 5% concentration of STKS, showing the lowest absorption and compressive strength but the highest density (Nurfitasari 2018).

In addition to empty palm bunches, Wicaksono et al. (2021) used nanocellulose as a filling material in the making of tapioca biofoam. Nanocellulose was obtained from manioc bark using the acid hydrolysis method with $H_2SO_4$ 6.5 M for 20 minutes. The size of the nanocellulose obtained ranged from 20-150 nm. The results showed that the use of nanocellulose fiber proportion increased the thickness of bioplastics and inhibited light in UV light and visible light (Wicaksono et al. 2021).

Khalid et al. (2020) also conducted physical properties testing on biopolymer with the addition of NC, using nanocellulose as a viscosity enhancement agent in polymer injection. Nanocellulose was obtained from cotton leaves through the ultrasonication process using an ultrasonic bath for 1 hour. The results showed that KLNC (cotton leaf nanocellulose) increased the viscosity of water, thereby increasing the effectiveness and efficiency of the enhanced oil recovery method in the petroleum industry (Khalid et al. 2020). Ghasemi et al. (2018) used nanocellulose together with polylactic acid (PLA) to make biocomposites. The results of mixing the NCF with maleated PLA increased the characteristic natures of the tensile modulus by 40% and the tensile strength increased from 22.4 MPa to 69.3 MPa from the addition of 5% (w/w) NCF (Ghasemi et al. 2018).

In a study conducted by Ramesh and Radhakrishnan (2019), adding cellulose nanofibrils (CNF) from potato peelings to PVA biopolymers improved the characteristics of active packaging. With the addition of CNF, the characteristics of PVA-CNF nanocomposite increased the value of tensile strength and elongation. Therefore, biopolymer active packaging based on the addition of CNF became more elastic and pliable (Ramesh and Radhakrishnan 2019).

From the studies mentioned above, it can be concluded that nanocellulose has significant potential as a biopolymer. The addition of nanocellulose in various studies has shown improvements in the physical characteristics of the materials. It enhances tensile strength, elongation, tensile modulus, and viscosity, while also increasing the thickness of modified composites. Nanocellulose is a promising additive in polymer matrices due to its renewable and biodegradable nature, as well as its abundance in nature. However, it is important to note that the optimal concentration of nanocellulose in film formulas has certain limits, as higher concentrations may negatively impact the mechanical properties and heat stability of the composite film (Owolabi et al. 2020; Abral et al. 2020; Ahankari et al. 2021). Therefore, further studies are required to explore these effects in more detail.

**Application in medical and cosmetic**

Fikri (2017) utilized nanocellulose derived from sweet potato leaves with a carbohydrate content of 8.82% through a 45-minute hydrolysis acid method using $H_2SO_4$ 45%. This nanocellulose was used in the production of gel masks. Similarly, Bongao et al. (2020) employed nanocellulose from pili pulp as an anti-aging ingredient in cosmetics, obtained through hydrolysis acid method (Bongao et al. 2020).

**Table 12**. Application of nanocellulose in medical and cosmetic fields

| Application of Nanocellulose | Material | Result | Reference |
|---|---|---|---|
| Gel face mask | Red sweet potato leaf (nanocellulose) | The tensile strength increases with the addition of nanocellulose | (Fikri 2017) |
| Anti-aging in cosmetics | Pili pulp (nanocellulose) | The optical nature of cellulose suspensions is almost equal to that of conventional mineral-based nanoparticles used in cosmetic | (Bongao et al. 2020) |
| Drug conductor for antimicrobial wound dressing | CNC medical cotton, polyvinylpyrrolidone (PVP) | The concentration of the drug slowly increases and remains constant after 48 hours | (Taher et al. 2020) |
| Alginate hydrogel as a drug conductor | Magnetic nanocellulose (m-CNCs) from CNC rice husk | Magnetic nanocellulose improves the physical and mechanical properties of alginate hydrogel, increases swelling, and decreases the rate of drug release | (Supramaniam et al. 2018) |
| Drug-conducting composite hydrogel for cancer therapy | Agrosa, CNC, polydopamine (PDA) | Compared to a single tissue hydrogel, the composite hydrogel achieves drug release of up to 86.3% through sustained release for 8 days at a pH of 5.5 | (Ning et al. 2021) |

Nanocellulose also finds applications in the healthcare sector. Taher et al. (2020) employed a mixture of nanocrystal cellulose (CNC) and polyvinylpyrrolidone (PVP) as a drug conductor for antimicrobial wound dressings in the form of honey. CNC was obtained from the synthesis of medical cotton using a 1M NaOH alkaline solvent method at a temperature of 160 °C for 2 hours. The release of honey from the film followed first-order kinetics, with a sustained release of the active ingredient over 48 hours. The nanocellulose film also exhibited significant antimicrobial activity against both gram-positive and gram-negative bacteria (Taher et al. 2020).

Then, Supramaniam et al. (2018) using magnetic nanocellulose (m-CNCs) synthesized from cellulose nanocrystals (CNCs) from rice husks in the making of alginate hydrogel. CNCs is synthesized using the acid hydrolysis method. The results show that magnetic nanocellulose improves the physical and mechanical properties of alginate hydrogels, increases the swelling and decreases the rate of drug release (Supramaniam et al. 2018).

Besides Ning et al. (2021) also uses cellulose nanocrystals (CNC) as the hydrogel composite material for drug-conducting paclitaxel (PTX) towards cancer therapy. CNC is combined with agrosa and polydopamine (PDA). Compared to single tissue hydrogel, Composite hydrogel produce drug release up to 86.3% through sustained drug release for 8 days at a pH of 5.5 (Ning et al. 2021).

From some of the above applications, nanocellulose is able to increase the tensile strength properties in the cosmetic field. Furthermore, nanocellulose is also able to reduce the release rate of drugs in the cosmetic field.

**Application in fuel and energy storage**

The bioconversion of lignocellulosic biomass into renewable biomass-based fuels has garnered significant attention in recent years. The current market demands necessitate the development of environmentally friendly products derived from new and renewable resources. The production of bioethanol from lignocellulosic biomass offers numerous advantages in terms of environmental impact and energy sustainability. Biomass is an economically viable raw material that is considered sustainable and renewable. It serves as a potential alternative to fossil-derived products, particularly in the energy sector, encompassing heat generation, fuel production, materials, and chemical compounds (Menon and Rao 2012).

Among the various pretreatment methods, the acid method employing $H_2SO_4$ is widely practiced due to its effectiveness in dissolving the holocellulose content while preserving the CNC integrity (Usov et al.

2015). Consequently, many industries utilize this method due to its inclination towards green processes (environmentally friendly processes) that enable the reuse of the acid solvent for the holocellulose content, renewal of the utilized materials, and separation of degraded sugar content for subsequent biofuel production via fermentation (Klemm et al. 2018).

**Table 13**. Application of nanocellulose in fuel

| Application of Nanocellulose | Material | Result | Reference |
|---|---|---|---|
| Bioethanol | Palm trunk | Nanocellulose derived from biomass in the form of palm trunks, rubber wood, and other hardwood mixtures has the potential to produce ethanol with conversions of 66%, 57%, and 60%, respectively. | (Adojo and Setyawan 2018) |
| BC Nanofiber with derived materials as electrodes in various energy storage devices | Bacterial nanocellulose (BNC) | Replacing conventional electrodes with BC Nanofiber-based electrodes can increase the capacity of lithium-ion batteries and improve battery cycle stability | (Guo et al. 2020) |
| Bioethanol | Biomass formosan alder | Among the various pretreatment methods used for producing bioethanol and nanocrystal cellulose (CNC), the best results were obtained using the Filter Paper (FP) Method pretreatment, yielding 383.4 mg glucose per gram of biomass, with a sugar fermentation yield of 4.18 g bioethanol per gram of biomass, and a CNC crystallinity of 613.93 nm | (Ko et al. 2020) |
| Biofuel | Whatman filter paper | Biofuel conversion in cellulose II (amorphous cellulose from hydrolysis waste) is 30% higher than in cellulose I (original cellulose) | (Pirani and Hashaikeh 2013) |

Admojo and Setyawan (2018) investigated the utilization of nanocellulose for bioethanol production. Nanocellulose derived from biomass sources such as palm trunks, rubber wood, and other hardwood mixtures exhibited ethanol conversion rates of 66%, 57%, and 60%, respectively (Admojo and Setyawan 2018).

Furthermore, Guo et al. (2020) explored the application of bacterial cellulose (BC) as an electrode material in various energy storage devices, including batteries. Substituting conventional electrodes with BC nanofiber-based electrodes demonstrated increased capacity in lithium-ion batteries and enhanced battery cycle stability (Guo et al. 2020).

## CONCLUSION

Among the various methods of nanocellulose synthesis, both the ionic liquid and DES (Deep Eutectic Solvent) methods offer environmentally safe waste and demonstrate superior temperature, time, and diameter control compared to other methods. However, the DES method has gained more prominence in recent times over the ionic liquid method due to the possibility of separating lignin waste from the DES solvent. The extracted lignin can be utilized in the production of fuel and aromatic compounds, while the solvent can be reused. Nanocellulose finds application in diverse fields such as film production, polymer science, cosmetics, medicine, fuel, and energy storage, where it enhances physical and mechanical characteristics.

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